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THE FORMATION OF ALLANITE-(Ce) IN CALCIC GRANOFELSES, NAMAQUALAND, SOUTH AFRICA

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ABSTRACT

Significant concentrations of allanite-(Ce) are reported from two calcic granofelses in the granulite facies of the western Namaqualand Metamorphic Complex, South Africa. The host rocks have plagioclase–garnet–clinopyroxene–quartz–titanite parageneses and are associated with mid-Proterozoic supracrustal quartzofeldspathic gneisses and quartz–biotite–sillimanite schists. Allanite-(Ce) occurs in two nonmetamict forms: firstly in a plagioclase–quartz gneiss as bands of prismatic grains with relatively high *REE* contents and simple zoning to a Ca-, Al-rich rim, and secondly with lower *REE* contents as coarse xenoblasts and fine inclusions in sieve-textured poikiloblasts in massive plagioclase–garnet granofelses. The coarse xenoblasts display reverse zoning to a Ca-, Al-rich core. The allanite-(Ce) concentrations in the banded plagioclase–quartz gneiss are considered to represent premetamorphic detrital accumulations derived from the weathering of felsic volcanic rocks. *REE* concentrations in the plagioclase–garnet granofelses represent metamorphic mobilisates into adjacent impure carbonate layers, that were initially accommodated in epidote at low metamorphic grades and subsequently in residual allanite-(Ce) enclaves when *P–T* conditions exceeded the stability range of epidote.

Keywords: allanite-(Ce), mineral composition, metamorphic reaction, calcic granofels, granulite facies, Namaqualand, South Africa.

SOMMAIRE

Des concentrations importantes d'allanite-(Ce) ont été découvertes dans deux granofels calciques du socle métamorphique (facies granulite) du Namaqualand occidental, en Afrique du Sud. Les roches encaissantes, qui contiennent l'assemblage plagioclase - grenat - clinopyroxène - quartz - titanite, sont associées à une séquence supracrustale d'âge protérozoïque moyen contenant des gneiss quartzo-feldspathiques et des schistes à quartz + biotite + sillimanite. L'allanite-(Ce) s'y trouve sous deux formes non métamictes. Dans un gneiss à plagioclase + quartz, elle forme des passées de grains prismatiques ayant des teneurs élevées en terres rares et une zonation simple vers une bordure enrichie en Ca et Al. Une allanite moins enrichie en terres rares est présente sous forme de xénoblastes grossiers et d'inclusions fines dans des poeciloblastes à texture de tamis dans des granofels massifs à plagioclase + grenat. Les xénoblastes grossiers montrent une zonation inverse, vers un coeur enrichi en Ca et Al. Les concentrations d'allanite-(Ce) dans les gneiss à plagioclase + quartz représenteraient des accumulations détritiques, et donc prémétamorphiques, dérivées d'un socle volcanique felsique lessivé. Les concentrations de terres rares dans les granofels à plagioclase + grenat résulteraient de leur mobilisation métamorphique dans des niveaux carbonatés voisins; les terres rares, situées dans l'épidote à un faible degré de métamorphisme, ont par la suite été concentrées dans des enclaves d'allanite-(Ce) quand les conditions de *P* et de *T* ont dépassé le champ de stabilité de l'épidote.

(Traduit par la Rédaction)

Mots-clés: allanite-(Ce), composition chimique, réaction métamorphique, granofels calcique, facies granulite, Namaqualand, Afrique du Sud.

INTRODUCTION

Allanite-(Ce) is reported as a minor to trace constituent of metamorphosed carbonate rocks (Deer *et al.* 1986), commonly from skarn assemblages at contacts with felsic igneous rocks (Papunen & Lindsjö 1972, Pavelescu & Pavelescu 1972, Derrick 1977, Gieré 1986), and more rarely from regionally metamorphosed calc-silicate assemblages (Sargent 1964). At the majority of these localities, allanite-(Ce) occurs in association with mineral parageneses that include calcic plagioclase, clinopyroxene, grandite garnet, scapolite, calcic amphiboles and titanite.

The chemical relationship between allanite and epidote is governed by the coupled replacement of Ca^{2+} + Fe^{3+} (epidote) for REE^{3+} + Fe^{2+} (allanite). In the epidote structure, Ca occupies two crystallographic sites of different sizes; the light rare-earth elements show a preference for the larger 10-fold coordinated *A*2 site (Dollase 1971). Allanite by definition contains in excess of 50% rare-earth elements (*REE*) in the *A*2 site, equivalent to a *REE* oxide content of about 15 wt.%. In nature, a complete range of *REE* contents is found from epidote to allanite (Maaskant *et al.* 1980, Sakai *et al.* 1984, Deer *et al.* 1986, Gieré 1986).

Epidote and clinozoisite are common constituents of impure carbonate metasediments at low amphibolite grades of metamorphism. With increase in metamorphic grades to temperatures between 600 and 700°C in the pressure range 3 - 5 kbars, epidote breaks down to form mineral assemblages that include anorthite, grandite garnet and magnetite (Holdaway 1972, Liou 1973). Epidote in metasedimentary rocks may contain significant amounts of the *REE* (Maaskant *et al.* 1980), and detrital allanite-(Ce) is reported from certain metapelitic rocks, rimmed by *REE*-bearing metamorphic epidote (Sakai *et al.* 1984).

Epidote is widely distributed in the western Namaqualand Metamorphic Complex (NMC), where it is particularly prevalent in amphibolites and calc-silicate rocks in the amphibolite-facies terrane (Coetzee 1941, Moore 1977) and as a minor retrograde constituent of mafic granulites and plagioclase-rich gneisses in the granulite-facies terrane (Joubert 1971). Allanite-(Ce) occurs in local stratabound concentrations in association with tourmaline, apatite, monazite, zircon and magnetite in leucocratic quartzofeldspathic gneisses in the Kakamas area of the western NMC (Hugo 1961). Minor metamict allanite-(Ce) also is described from simple pegmatites in the Onseepkans–Kenhardt pegmatite belt (Hugo 1970) and from alaskitic leucogranitic bodies in the Springbok area (Robb 1986).

We report here the presence of significant concentrations of allanite-(Ce) (hereafter referred to as allanite) in certain calc-silicate rocks in the granulite-facies portion of the western NMC. These occurrences provide textural and chemical evidence that has a direct bearing on the formation of allanite in calc-silicate rocks, and leads to proposals concerning

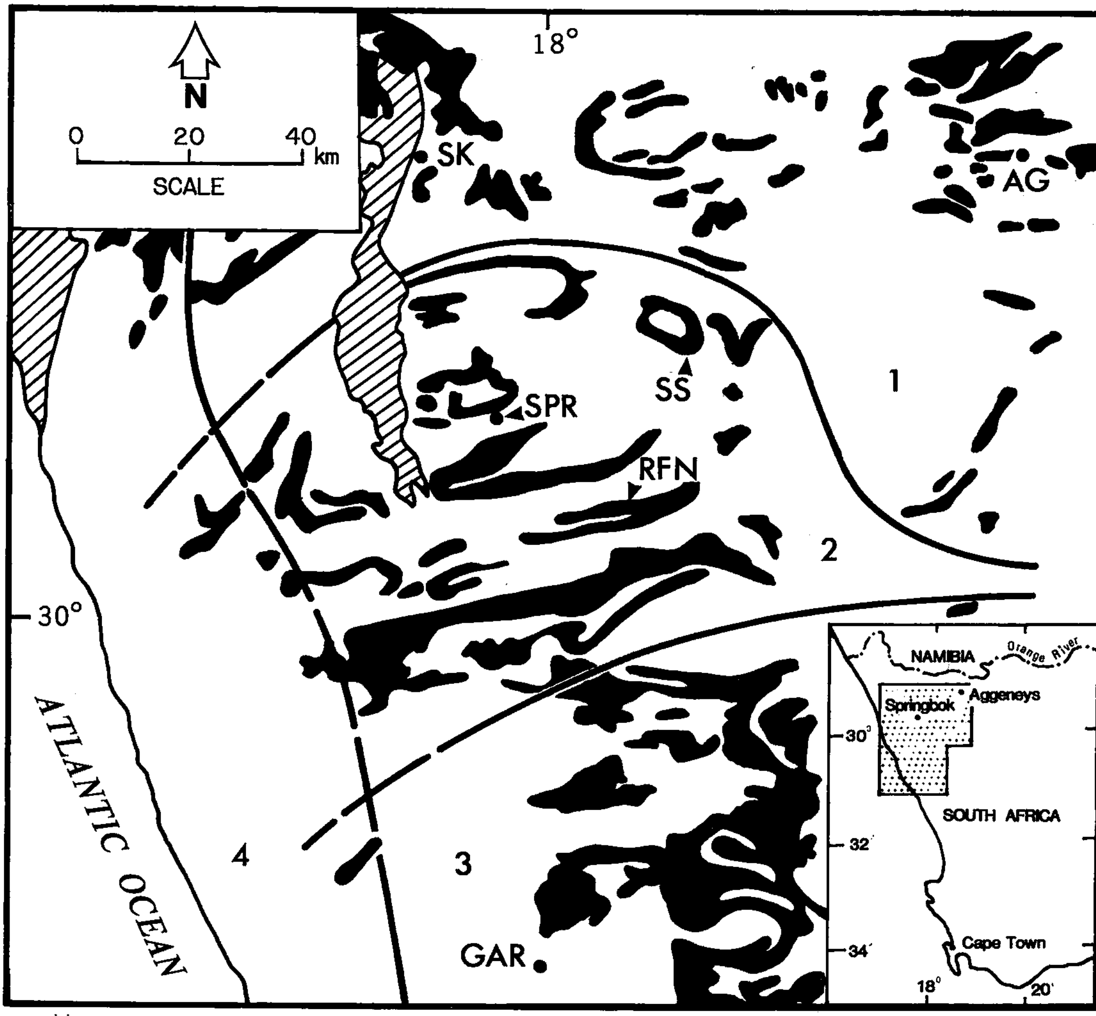

FIG. 1. Regional map of portion of the western Namaqualand Metamorphic Complex, showing the distribution of supracrustal rocks (black), granitic gneisses (unadorned) and overlying Pan-African metasediments (hatching). The regional metamorphic zonation includes amphibolite facies (1), cordierite–garnet subzone of the granulite facies (2), spinel–quartz subzone of the granulite facies (3) of the Namaqua event, and overprinted Pan-African staurolite zone (4). Localities are Smorgen Schaduwe (SS) and Rietfontein (RFN), and towns are Garies (GAR), Springbok (SPR), Aggeneys (AG) and Steinkopf (SK). Inset shows location of map area.

the origins of high concentrations of light-rare-earth minerals in certain mid-Proterozoic metamorphosed carbonate rocks.

General Geology

The allanite occurrences are located on the farms Smorgen Schaduwe and Rietfontein, respectively 45 km east and 40 km southeast of Springbok, Namaqualand, South Africa (Fig. 1). The calc-silicate lenses that host the allanite concentrations occur within remnants of a supracrustal volcano-sedimentary sequence in a terrane dominated by intrusive granitic and charnockitic augen gneisses (Joubert 1971, Moore 1986). At Rietfontein, the supracrustal sequence comprises quartzofeldspathic biotite gneisses and leucogneisses, with minor calc-silicate, basic granulite and amphibolite bands.

These lithologies are typical of the basal portions of the supracrustal tectonostratigraphic sequence in western Namaqualand (Moore 1986). In contrast, the sequence at Smorgen Schaduwe is dominated by quartz–biotite–sillimanite schists and quartzites, with minor garnetiferous iron formations, typical of the upper stratigraphy of the supracrustal sequence. Amphibolites in this portion of the stratigraphy at Aggeneys, 100 km to the east, have yielded a Sm–Nd isochron age of 1650 Ma (Reid *et al.* 1987).

Metamorphic zonation in the western NMC (Fig. 1) is chiefly defined by mineral assemblages in metapelitic rocks. The paragenesis biotite + sillimanite + quartz typifies the upper amphibolite facies, with P–T estimates in the region of 650°C at 5 kbars; cordierite + garnet + K-feldspar + quartz typifies the lower-T portion of the granulite facies (750°C at 5 kbars), and hercynite + quartz, the high-T portion of the granulite facies (>800°C at 5 kbars) (Waters 1989). The two occurrences of allanite fall within the low-T granulite-facies terrane (Fig.1).

At the Bobbejaanpoort locality on the farm Rietfontein, allanite-rich rocks occur as boudinaged pods of dark brown granofels in an impersistent band, 10–20 cm thick and 100 m long, enclosed within a 10–20-m-thick calc-silicate unit. The allanite-rich pods consist of a thin (1–5 cm), finer-grained plagioclase–allanite–magnetite–titanite layer, directly overlain by a thicker (10–15 cm), coarser-grained plagioclase–garnet–clinopyroxene–allanite layer that grades progressively into an allanite-free granofels of otherwise similar composition. The enclosing calc-silicate assemblages are both garnet granofelses and clinopyroxene-bearing rocks, and are themselves enclosed by magnetite-rich quartzofeldspathic gneisses. Several other calc-silicate units are present within the quartzofeldspathic gneisses on Rietfontein, but none of these have yielded allanite concentrations.

TABLE 1. BULK COMPOSITIONS OF ALLANITE-(Ce)-BEARING AND ASSOCIATED ROCKS FROM SMORGEN SCHADUWE AND RIETFONTEIN

	1.	2.	3.	4.	5.
SiO_2 (wt.%)	44.54	60.01	40.03	92.66	60.14
TiO_2	.48	.66	.46	.04	.12
Al_2O_3	21.19	17.38	20.12	3.29	8.11
FeO*	9.67	5.65	12.49	.89	6.51
MnO	.17	.13	.31	.05	.24
MgO	.19	.49	.20	.71	7.10
CaO	19.81	7.59	19.40	2.40	15.04
Na_2O	.56	4.80	.75	.23	2.53
K_2O	.07	.30	.04	.05	.36
P_2O_5	<.02	.21	.21	.03	.49
LOI	.43	.90	1.85	.19	.13
H_2O_-	.04	.16	.09	.10	.09
TOTAL	97.15	98.28	95.95	100.64	100.86
Rb (ppm)	20	7	<1	4	15
Sr	1440	557	586	33	228
Th	61	118	23	<5	13
Zr	213	346	226	41	78
Nb	99	50	38	<5	3
Y	31	299	210	4	40
La	4785	4328	7608	5	42
Ce	8090	7447	11309	7	107
Nd	1935	1871	2589	4	56

*total Fe as FeO. LOI – loss on ignition. 1. SS-4, massive garnet-clinopyroxene-allanite-(Ce)-plagioclase rock, Smorgen Schaduwe. 2. SS-18, banded plagioclase-quartz-allanite-(Ce)-titanite rock, Smorgen Schaduwe. 3. RFN-20, massive garnet-clinopyroxene-allanite-(Ce)-plagioclase rock, Rietfontein. 4. SS-19, quartz-clinopyroxene calc-silicate rock, Smorgen Schaduwe. 5. SS-3, plagioclase-clinopyroxene calc-silicate rock, Smorgen Schaduwe.

Analyses by Philips PW1400 and Siemens SRS-1 automatic wavelength-dispersive sequential X-ray fluorescence spectrometers in the Geochemistry Department, University of Cape Town. For operating procedures and conditions see Moore (1986).

At Smorgen Schaduwe, allanite-bearing rocks occur locally at the contact between quartzofeldspathic gneisses and an overlying sequence of quartz–biotite–sillimanite schists and quartzites. The allanite-rich rocks consist of a lower unit (± 1 meter thick) of plagioclase–quartz gneiss containing discrete bands of clinopyroxene, allanite, titanite and magnetite, and overlying pods (20–30 cm thick) of more massive garnet–clinopyroxene–allanite–plagioclase rock, similar to those at Rietfontein. The allanite-rich calc-silicate rocks are directly overlain by clinopyroxene–hornblende–quartz granofelses that become increasingly quartzitic away from the allanite band.

The bulk chemical compositions of the allanite-rich calc-silicate rocks reveal that the rocks are generally calcic (20 wt.% CaO), magnesium-poor (<0.50 wt.% MgO), peraluminous (20 wt.% Al_2O_3) and iron-rich (10 wt.% FeO), containing approximately 2 wt.% *REE* oxides (Table 1). The plagioclase–quartz gneiss at Smorgen Schaduwe is, in addition, highly sodic (5 wt.% Na_2O). Relatively high levels of strontium (up to 1500 ppm) are present in all rock types. Enrichment in the *REE* is not associated with significant concentrations in other incompatible elements such as K, Rb, Zr, Th, Nb and P. Overlying clinopyroxene–quartz granofelses at Smorgen Schaduwe are more magnesium-rich, sodic (2.5 wt.% Na_2O) and have dramatically lower *REE* contents (< 500 ppm, Table 1).

TABLE 2. CHEMICAL COMPOSITION OF GRANDITE GARNET, CLINOPYROXENE AND TITANITE FORM SMORGEN SCHADUWE AND RIETFONTEIN

	1.	2.	3.	4.
SiO_2 (wt.%)	35.97	46.99	45.42	29.90
TiO_2	0.65	0.18	0.33	26.83
Al_2O_3	6.95	3.61	4.05	4.70
Fe_2O_3	20.97			
FeO	3.62	20.99*	23.68*	3.96*
MnO	0.51	0.37	0.77	0.12
MgO	0.11	4.93	3.18	0.13
CaO	31.55	22.37	22.17	26.40
Na_2O	-	0.60	0.53	-
La_2O_3	-	-	-	0.30
Ce_2O_3	-	-	-	1.32
Nd_2O_3	-	-	-	0.79
Pr_2O_3	-	-	-	0.21
Sm_2O_3	-	-	-	0.15
Y_2O_3	-	-	-	0.59
F	-	-	-	2.25
TOTAL	100.33	100.04	100.13	97.65
O	12	6	6	20
Si	2.941	1.870	1.836	4.112
Ti	0.040	0.005	0.010	2.775
Al	0.671	0.169	0.193	0.762
Fe^{3+v}	1.290			
Fe^{2+}	0.247	0.698	0.800	0.455
Mn	0.035	0.012	0.046	0.013
Mg	0.013	0.292	0.192	0.026
Ca	2.763	0.954	0.960	3.890
Na	-	0.077	0.042	-
La	-	-	-	0.015
Ce	-	-	-	0.066
Nd	-	-	-	0.038
Pr	-	-	-	0.010
Sm	-	-	-	0.007
Y	-	-	-	0.043
F	-	-	-	0.489
SUM	8.000	4.077	4.079	12.701

v Fe^{3+} estimated after Droop (1987). * total Fe as FeO.
1. Grandite garnet, sample SS-4, Smorgen Schaduwe. 2. Clinopyroxene sample SS-4, Smorgen Schaduwe. 3. Clinopyroxene, sample McS-351, Rietfontein. 4. Titanite, sample SS-18, Smorgen Schaduwe.
For analytical procedure see Table 3.

Petrography and Mineral Chemistry

Plagioclase is the major constituent (38–73 vol.%) of all the allanite-bearing lithologies and commonly occurs in granoblastic mosaics of equidimensional grains. It is mostly unzoned and generally contains numerous inclusions of quartz, allanite, apatite and zircon. Plagioclase shows considerable compositional variation, from An_{80-87} at Rietfontein to An_{77} in the allanite-rich pods and An_{36} in the plagioclase–quartz gneiss at Smorgen Schaduwe. Scapolite (Me_{77}) is associated with plagioclase in the allanite-rich pods at Smorgen Schaduwe.

Garnet occurs in all the allanite-bearing rocks (up to 22 vol.%), although it is a rare constituent in the plagioclase–quartz gneiss. It is generally present as large dendritic poikiloblasts enclosing allanite and plagioclase. Garnet locally forms a corona enclosing clinopyroxene and also occurs in complex symplectite with magnetite and quartz, replacing clinopyroxene. Garnet compositions from the various lithologies have restricted ranges: $andradite_{60-55}$ $grossular_{40}$ $almandine_{0-5}$ (Table 2).

Clinopyroxene is present as dark green polygonal grains (2–13 vol.%) containing, in places, fine inclusions of rutile. It has a composition in the hedenbergite field and a low Na content (Table 2). In contrast to plagioclase and garnet, clinopyroxene contains few inclusions of allanite.

Allanite (10–33 vol.%) shows several textural variations within the calcic granofelses. In the allanite-rich pods, it commonly occurs as coarse (1 mm), brown to red-brown, xenoblastic grains showing simple twinning. The allanite is nonmetamict, and clusters of grains show mutual 120° contacts (Fig. 2). A common additional texture observed in the allanite pods is a sieve-textured distribution of fine allanite grains in garnet and plagioclase (Fig. 3).

In the plagioclase–quartz gneiss at Smorgen Schaduwe and the thin plagioclase–allanite–magnetite–titanite layer at Rietfontein, allanite occurs in distinct bands as prismatic to tabular idioblastic grains generally elongated along the gneissic fabric (Fig. 4). Rarely, a complex intergrowth or symplectite between allanite and titanite is observed.

Allanite grains in the plagioclase–quartz gneiss at Smorgen Schaduwe show color zonation, with a darker core and distinct narrow (25 μm) pale rim (Fig. 4), corresponding to an increase in the epidote component (Fig. 5a). In the allanite-rich pods, the zoning is more complex, with large grains having a lighter core that grades progressively into a darker mantle which, in turn, reverts more abruptly to an impersistent light rim (Fig. 2). Both the paler core and rim are relatively enriched in Ca and Al (Fig. 5b).

The coupled substitution $Ca^{2+} + Fe^{3+} = REE^{3+} + Fe^{2+}$ is inadequate to account for charge balances in the zoned grains. It is apparent from Table 3 that the exchange mechanism requires complex substitutions in the *M*3 site, whereby both Fe^{3+} and Al^{3+} are replaced by Fe^{2+} and Mg^{2+} to maintain charge balance. Substitutions such as $Mg^{2+} + F^- = Al^{3+} + O^{2-}$ (Peacor & Dunn 1988) are not applicable, as F concentrations are below detection limit (0.24 wt.%) in the NMC allanite.

Allanite from the Namaqualand calcic granofelses has a low total *REE* content (0.50 to 0.76 in the *A*2 site) (Table 3). The rim of certain grains consists of allanitic epidote rather than allanite. The highest *REE* contents are recorded from the plagioclase–quartz gneiss at Smorgen Schaduwe, and the lowest, from the overlying allanite-rich pods. Allanite grains in the plagioclase–quartz gneiss contain higher Mg and Mn contents, whereas those from the allanite-rich pods are more Al-rich. The absence of detectable levels of Th and U accounts for the nonmetamict nature of the allanite.

Titanite is a minor constituent in all the allanite-bearing lithologies and is most common in the plagioclase–quartz gneiss at Smorgen Schaduwe, where it occurs as anhedral grains in layers with magnetite, diopside and allanite. The titanite shows considerable departure from the pure end-member, with complex substitutions involving Al and Fe for Ti, light *REE* and Y for Ca, and F for O (Table 2).

Quartz is present in the calcic granofelses either as disseminated xenoblastic grains or, at Rietfontein, as coarse veinlets rimmed by clinopyroxene that is being replaced by garnet–magnetite–quartz symplectite. Magnetite occurs as rounded anhedral grains, altering to a hematite rim, in the plagioclase–quartz gneiss at Smorgen Schaduwe and as arcuate plates in garnet symplectite in the allanite-rich pods. Small rounded zircon grains are observed in certain allanite bands in the plagioclase–quartz gneiss.

Formation of Allanite-(Ce)

Based on their bulk chemistry and local association with marble lenses, the calcic granofelses are believed to represent thin impure Fe-, Al-rich carbonate sedimentary layers associated with rhyolitic to rhyodacitic extrusive volcanic rocks (leucogneisses and biotite gneisses: Moore 1986) and overlying pelitic and siliceous sediments (quartz–biotite–sillimanite schists and quartzites). At amphibolite metamorphic grades, the carbonate sediments are represented by epidote-rich lithologies (Coetzee 1941, Moore 1977).

Epidote has a relatively wide field of stability in the temperature range 300 to 700°C at the pressures experienced in the western NMC (5 kbars) (Liou 1973, Liou *et al.* 1983). Grandite garnet has an even wider range in temperature at these pressures, forming initially in discontinuous reactions resulting in the breakdown of prehnite at temperatures between 350 and 400°C (Liou *et al.* 1983). Depending on epidote composition and prevailing $f(O_2)$ conditions, epidote breakdown occurs at temperatures between 600 and 700°C, forming garnet and calcic plagioclase (Liou 1973):

$$\underset{\text{epidote}}{36Ca_2FeAl_2Si_3O_{12}(OH)} = \underset{\text{anorthite}}{36CaAl_2Si_2O_8} +$$

$$\underset{\text{andradite}}{12Ca_3Fe_2Si_3O_8} + \underset{\text{magnetite}}{4Fe_3O_4} + O_2 + 18H_2O \qquad (1)$$

In the same temperature range, epidote will react with ilmenite to form titanite (thermodynamic calculations based on Powell & Holland 1988) in reactions such as:

$$\underset{\text{epidote}}{4Ca_2FeAl_2Si_3O_{12}(OH)} + \underset{\text{ilmenite}}{3FeTiO_3} + \underset{\text{quartz}}{SiO_2} =$$

$$\underset{\text{anorthite}}{4CaAl_2Si_2O_8} + \underset{\text{hedenbergite}}{CaFeSi_2O_6} +$$

$$\underset{\text{magnetite}}{2Fe_3O_4} + \underset{\text{titanite}}{3CaTiSiO_5} + 2H_2O \qquad (2)$$

Reactions 1 and 2 contain all the major constituents of the calcic granofelses of the western NMC, other than allanite.

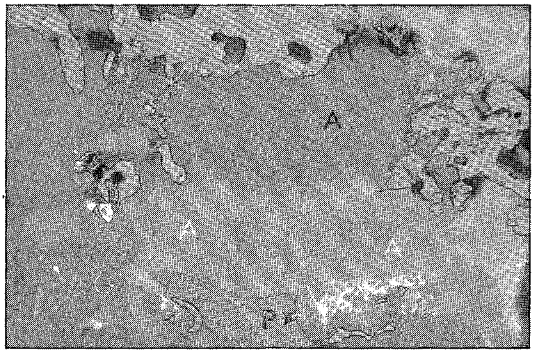

FIG. 2. Three grains of nonmetamict allanite-(Ce) (A) showing mutual 120° contacts and complex color zoning, with a paler core, a darker mantle and an impersistent pale rim. Also present are plagioclase (colorless), clinopyroxene (P) and grandite garnet (G). Sample SS-4, Smorgen Schaduwe. Photo length 2.5 mm.

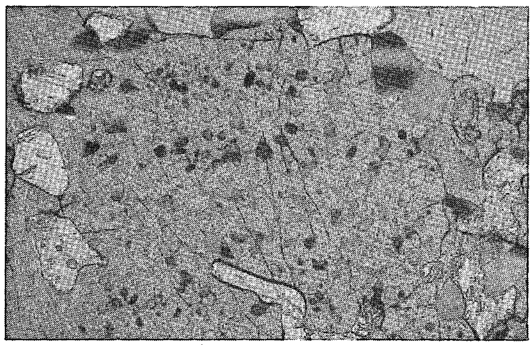

FIG. 3. Large grain of sieve-textured garnet poikiloblastically incorporating allanite-(Ce). Sample SS-4, Smorgen Schaduwe. Photo length 2.5 mm.

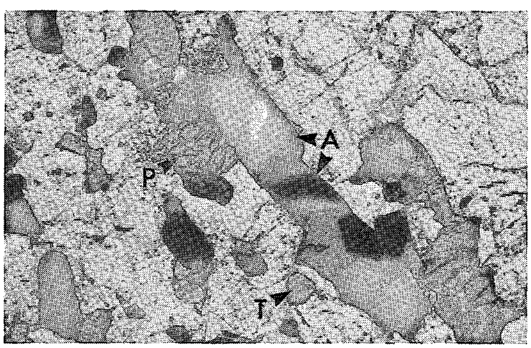

FIG. 4. Prismatic to tabular grains of allanite-(Ce) (A), each with a dark core and a thin pale rim, aligned in the fabric and associated with clinopyroxene (P), titanite (T) and plagioclase (colorless). Sample SS-18, Smorgen Schaduwe. Photo length 2.5 mm.

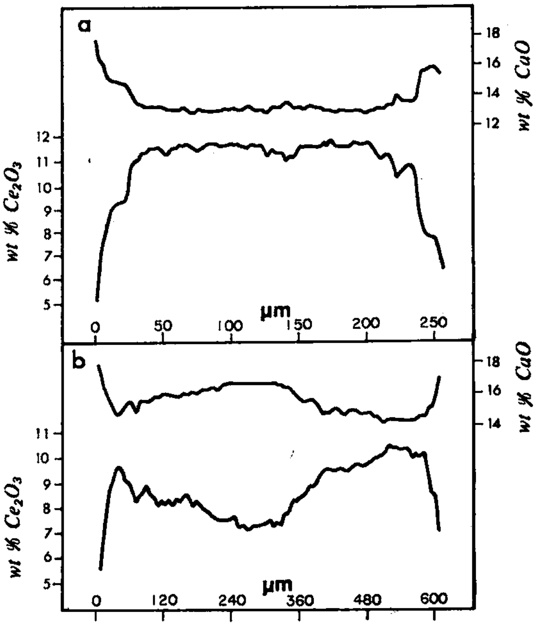

FIG. 5. a. Simple compositional zoning across a prismatic grain of allanite-(Ce), illustrating the Ce-poor, Ca-rich rim. Sample SS-18, Smorgen Schaduwe. b. Complex compositional zoning across large grain of allanite-(Ce), illustrating the Ce-poor, Ca-rich rim and core, and the Ce-rich, Ca-poor mantle. Sample SS-4, Smorgen Schaduwe.

In contrast to epidote, allanite is a stable mineral phase in the low-*T* portion of the granulite facies in the western NMC (~750°C). The upper stability limits of allanite in metamorphic terranes have yet to be reported, although critical saturation temperatures of approximately 800°C are reported for allanite crystallization in silicic magmas (Chesner & Ettlinger 1989). Assuming that the calcic granofelses contained *REE*-bearing epidote, then, on commencement of epidote breakdown during prograde metamorphism, the *REE* could not be accommodated by newly formed plagioclase, garnet and clinopyroxene. Instead they would be progessively concentrated in residual allanite enclaves. In the majority of calc-silicate rocks, only minor *REE* concentrations may be expected in epidote, resulting in the formation of allanite as an occasional trace constituent.

In the case of the calcic granofelses in the western NMC, however, significant *REE* concentrations (at least 2 wt.% *REE* oxides) must have been present in epidote to account for the large amount of allanite. The above mode of formation of allanite would explain its characteristic sieve-texture within plagioclase and garnet (Fig. 3). As epidote was progressively consumed in the breakdown reaction, small enclaves of increasingly *REE*-rich epidote and ultimately allanite became isolated within growing plagioclase and garnet. This process of allanite formation in addition accounts for the relatively low levels of the *REE* within allanite, as the *REE*-concentrating process would continue only until sufficient concentrations of the *REE* were accommodated within the epidote-group mineral to stabilize it at the prevailing *P–T* conditions of the granulite facies (~750°C, 5 kbars).

The narrow Ca-, Al-rich rim displayed by virtually all allanite grains formed in response to a post-peak cooling episode. The more complex pattern of zonation in large grains from the allanite pods (Fig. 2), however, represents a combination of the above process superimposed on an earlier reverse zonation. The latter zoning to a more Ca-, Al-rich core is a consequence of diffusion of the *REE* into the enclaves of residual allanite during epidote breakdown. Similar zoning to a Ca-, Al-rich core has been reported from metamorphic allanite in biotite-plagioclase-quartz hornfelses (Black 1970) and is associated with migration of the *REE* during metamorphism. Zonation in allanite of igneous origin is generally of the opposite sense, from *REE*-rich core to a Ca- and Al-rich rim (Morin 1977, Harding *et al.* 1982, Gromet & Silver 1983).

REE ENRICHMENT

The primary, but localized, concentrations of the *REE* in calcic granofelses of the western NMC are somewhat enigmatic, and several possible genetic models may be considered. The allanite-bearing granofelses generally show at least 1000× enhancements in light *REE* contents compared to similar garnet-plagioclase granofelses in the western NMC (Moore 1986) and immediately adjacent clinopyroxene granofelses (Table 1). Three different processes of enrichment, both pre- and syn-metamorphic, are considered below:

Detrital concentrations of REE-bearing minerals

The banded nature of the basal plagioclase-quartz gneiss at Smorgen Schaduwe and the plagioclase-allanite-magnetite-titanite layer at Rietfontein suggests that processes of sedimentary sorting may have been responsible for the concentration of detrital *REE*-bearing minerals as layers of heavy minerals in association with impure carbonate beds. If the layers do represent heavy-mineral sands, however, they must have been of an unusual composition, as modern-day *REE* placer deposits are dominated by the gangue minerals ilmenite, rutile and zircon and contain monazite as the principal *REE* ore (Neary & Highley 1984). Magnetite-, ilmenite-, and zircon-bearing heavy-mineral layers are reported from leucogneisses and quartzites elsewhere in the western NMC (Frick & Wheelock 1983, Lipson *et al.* 1986, Moore 1986).

Epidote occurs as one of the dominant heavy-mineral constituents in the Miocene diamondiferous gravels of the Orange River (van Wyk & Pienaar 1986). It is conceivable that allanite may be selectively concentrated during the weathering of felsic volcanic rocks. Allanite commonly occurs as a minor constituent of felsic to intermediate igneous rocks (Ghent 1972, Morin 1977, Brooks *et al.* 1981, Harding *et al.* 1982, Gromet & Silver 1983, Mitropoulos 1987, Chesner & Ettlinger 1989). Allanite in the plagioclase–quartz gneiss has the highest *REE* content (Table 3) and consists of anhedral grains that lack both the sieve texture and complex reverse zonation patterns of the allanite-rich pods. Titanite (after ilmenite, see reaction 2), magnetite and zircon (350 ppm Zr) have their highest concentrations in this gneiss (Table 1).

TABLE 3. CHEMICAL COMPOSITION OF ALLANITE-(Ce) FROM SMORGEN SCHADUWE

	1.	2.	3.	4.	5.
SiO_2 (wt.%)	32.73	32.09	33.31	31.60	32.81
TiO_2	0.33	0.30	0.26	0.43	0.27
Al_2O_3	16.96	16.15	17.56	14.59	16.40
Fe_2O_3*	10.21	9.30	10.83	7.76	10.36
FeO	5.39	6.59	4.58	9.46	6.20
MnO	nd	nd	nd	0.28	0.29
MgO	0.30	0.34	0.23	0.51	0.38
CaO	15.77	14.35	16.93	13.03	15.04
La_2O_3	4.82	5.89	4.19	6.59	5.10
Ce_2O_3	8.86	10.27	7.38	11.36	8.89
Nd_2O_3	1.45	1.58	1.16	1.90	1.60
Pr_2O_3	0.65	0.76	0.53	0.85	0.69
TOTAL	97.47	97.62	96.96	98.36	98.03
O	25	25	25	25	25
Si	5.951	5.942	5.953	5.920	5.967
Ti	0.022	0.021	0.035	0.061	0.037
Al	3.635	3.526	3.700	3.223	3.516
Fe^{3+}	1.397	1.294	1.457	1.094	1.392
Fe^{2+}	0.820	1.020	0.685	1.481	0.925
Mn	–	–	–	0.044	0.045
Mg	0.081	0.094	0.061	0.142	0.103
Ca	3.072	2.848	3.242	2.615	2.931
La	0.323	0.402	0.276	0.456	0.342
Ce	0.590	0.697	0.483	0.779	0.592
Nd	0.094	0.105	0.074	0.127	0.104
Pr	0.015	0.051	0.034	0.058	0.046
SUM	16.000	16.000	16.000	16.000	16.000

*Fe^{3+} estimated after Droop (1987). nd – below detection limit.
1. Allanite-(Ce) core, sample SS-4, Smorgen Schaduwe.
2. Mantle of same grain. 3. Rim of same grain.
4. Allanite-(Ce) core, sample SS-18, Smorgen Schaduwe.
5. Rim of same grain.

Analysis by electron microprobe (Cameca, accelerating potential 25 kV, sample current 40 nA) against natural and synthetic mineral standards. REEs analysed using Drake & Weill (1972) standards, LiF crystal, Lα peaks for La, Ce, Y and Lβ peaks for Nd, Pr, Sm. PAP correction factors (Pouchou & Pichoir 1984) were applied.

Metamorphic mobilization of the REE

Under certain conditions, the *REE* are highly mobile at elevated temperatures, particularly in carbonatite, hydrothermal and low-*T* metamorphic systems (Wood *et al.* 1976, McLennan & Taylor 1979, Hellman *et al.* 1979, Andersen 1986, Gieré 1986). Mobility of the *REE* is largely dependent on the nature of the original mineral phases containing the *REE*, composition of the fluid phase, and ability of minerals formed during the reactions to accommodate the *REE* from the fluid (Humphris 1984). Fluorine–*REE* complexes in the fluid phase are considered to play a significant role in the migration of the *REE* (Alderton *et al.* 1980, Humphris 1984, Andersen 1986, Gieré 1986). Light-*REE* fractionation may occur during fluid transport owing to selective mobility of the *REE* (Hellman *et al.* 1979, Andersen 1986) or by selective accommodation in newly formed minerals.

During the initial low-*T* phase of the major prograde Namaqua metamorphism, it is conceivable that the *REE* were mobilized by leaching from minor mineral phases in the metavolcanic rocks and selectively accommodated in epidote that was forming in adjacent impure carbonate horizons. Minor amounts of F in titanite in the calcic granofelses suggest that F–*REE* complexes may have been present. If the *REE*-bearing fluids were not pervasive but restricted or channeled, this may explain the localized nature of the allanite occurrences.

CO_2-associated REE concentrations

The granulite-facies metamorphic conditions in the western NMC are closely associated with the intrusion of suites of charnockitic and anorthositic to noritic rocks (van Zyl 1978, McIver *et al.* 1983, Albat 1984, Andreoli & Hart 1987). Fractional crystallization of the anorthositic–noritic suite led to the residual concentration of silica-poor melts enriched in oxides (magnetite, ilmenite), sulfides (chalcopyrite, bornite, pyrite) and phosphates (apatite, monazite) (van Zyl 1978, Andreoli & Hart 1987). *REE* enrichment is apparent in these late-stage melts, as a result of partitioning of the *REE* into the residual melts and an associated CO_2 vapor phase (Wendlandt & Harrison 1979). Charnockitic veinlets enriched in lithophile elements, including the *REE*, are present in certain portions of the granulite facies of the western NMC, particularly in association with the anorthositic–noritic suite. These are believed to have formed at the peak of the granulite-facies metamorphism, when CO_2-rich vapors containing high concentrations of lithophile elements were released from the crystallizing H_2O-poor magmas and penetrated the country rocks (Andreoli & Hart 1987).

It is conceivable that such a vapor phase could have reacted locally with the calcic granofelses to form the allanite concentrations. The allanite-bearing rocks, however, do not contain the full complement of large-ion lithophile elements anticipated, with K, Th and P noticeably absent. Certain textural features (*REE* zonation pattern, sieve textures, lack of metamictization) indicate the presence of a precursor *REE*-bearing epidote mineral prior to the peak of metamorphism, when the deep-seated CO_2-rich vapor phase would not have been present.

Although the source of the *REE* in the calcic granofelses is by no means resolved, a model involving detrital concentration and localized low-*T* metamorphic remobilization is currently favored. Allanite may well have been a minor phase in the rhyolitic–rhyodacitic volcanic rocks, with local heavy-mineral bands forming during sedimentary reworking. With the onset of regional metamorphism, the *REE* were mobilized to a limited degree and selectively reconcentrated in adjacent epidote-rich calc-silicate layers. Increasing grades of metamorphism resulted in epidote breakdown and the formation of allanite of distinctive composition, zoning and texture in the calcic granofelses.

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